PB-231 256

MERCURY IN THE ENVIRONMENT

David H. Klein

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Prepared for:
Environmental Protection Agency

December 1973

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PB 231 256

SELECTED WATER RESOURCES ABSTRACTS
INPUT TRANSACTION FORM

1. Report No. | 2. | W 74-06523

MERCURY IN THE ENVIRONMENT,

5. Report Date
6.
8. Performing Organization Report No.

Klein, D. H.

EPA Project 16040 FRL

Hope College, Holland, Michigan, Chemistry Department

13. Type of Report and Period Covered

~~12. Sponsoring Organization Environmental Protection Agency~~

Environmental Protection Agency, Technology Series Report ~~Number~~ EPA-660/2-73-008, December 1973. 23 p, 6 tab, 9 ref. src presented

~~This report presents the~~ results of studies of dilute discharges of mercury and some other metals into the characteristically low-metals environment of western Michigan, together with information on the transport of some of these metals through the environment, and on their sinks in the sediments. Atmospheric discharge sources include power plants, airports, asphalt plants, and general urban industrialization. Metals less volatile than mercury are deposited in surface soils near the point of discharge, but only a small portion of the mercury is deposited near the point of discharge. Sewage treatment plants represent a source of discharge of fairly large amounts of mercury - about 1000 lb per million population. This mercury is associated primarily with the suspended solids. In local lakes and rivers, mercury is also associated primarily with the suspended solids. In sediments, the concentrations of mercury and other metals correlate strongly with the organic content and the fraction of fine particulates. (Alford-EPA)

17a Descriptors

*Mercury, *Metals, *Water pollution, *Air pollution, *Water pollution sources, Soil contamination, Sediments, Suspended solids, Path of pollutants, *Michigan, Treatment facilities

17b Identifiers

*Air pollution sources, ~~southwestern Michigan, lakes and rivers in Michigan,~~ sewage treatment plants, coal-burning power plants

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17c. COWRR Field & Group 05B

Copy Available from GPO Sup Doc as EP1.23:660/2-73-008, $0.65; microfiche from NTIS as PB $1.45

18. Availability

19. Security Class. (Report)
20. Security Class. (Page)
21. No. of Pages
22. Price

Send To:
WATER RESOURCES SCIENTIFIC INFORMATION CENTER
U.S. DEPARTMENT OF THE INTERIOR
WASHINGTON, D. C. 20240

Abstractor Ann L. Alford | Institution SERL, Athens, Georgia 30601

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EPA-660/2-73-008
December 1973

MERCURY IN THE ENVIRONMENT

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Program Element 1BA027

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ABSTRACT

This report presents the results of studies of dilute discharges of mercury and some other metals into the characteristically low-metals environment of western Michigan, together with information on the transport of some of these metals through the environment, and on their sinks in the sediments.

Atmospheric discharge sources include power plants, airports, asphalt plants, and general urban industrialization. Metals less volatile than mercury are deposited in surface soils near the point of discharge, but only a small portion of the mercury is deposited near the point of discharge.

Sewage treatment plants represent a source of discharge of fairly large amounts of mercury - about 1000 lb per million population. This mercury is associated primarily with the suspended solids.

In local lakes and rivers, mercury is also associated primarily with the suspended solids.

In sediments, the concentrations of mercury and other metals correlate strongly with the organic content and the fraction of fine particulates.

This report was submitted in fulfillment of Project Number 16040 FRL, by Hope College, under the partial sponsorship of the Environmental Protection Agency. Work was completed as of May 1973.

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SECTION I

CONCLUSIONS

There exist a number of sources of discharge of mercury to the environment which, though dilute, are sufficient to create an imprint in soils and sediments. Such imprints resulting from atmospheric discharge have been observed in an urban industrial area. Specific sources of mercury discharge include airports, asphalt plants, and most significantly, coal burning power plants.

Other metals discharged from power plant stacks travel only a few miles before falling to the ground, but mercury is discharged as a vapor and is only slightly enriched in nearby soils. Most of the mercury discharged apparently enters an atmospheric circulation pattern different from that of the less volatile metals.

The complexities of sediment transport systems have prevented detection of a similar imprint in the low-mercury sediments characteristic of western Michigan. However, mercury and a number of other elements show a strong tendency to be associated with the organic fraction of the sediments.

The low levels of mercury found in western Michigan waters are associated primarily with the suspended solids, which have mercury concentrations of about 10 ppm. Similar concentrations are found on the suspended solids discharged from a sewage treatment plant. Contact of waste water with large quantities of such mercury-loaded sludges sometimes results in a desorption of mercury from the sludge into the effluent.

SECTION II

RECOMMENDATIONS

Mercury from burning coal is dispersed widely, and may enter the aquatic or terrestrial environment far from the point of discharge. Since mercury discharged in this way is of the same order of magnitude as the total of mercury mined in the world, it appears advisable to try to develop a technology to remove mercury either from the coals or from stack gases. Some effort should also be expended to locate and study lakes in which airborne mercury has made an environmental impact.

Mercury and other toxic metals in sediments are associated primarily with the organic fraction of the sediments. Decreasing BOD loadings to waters, especially of organic particulates, should act to decrease the mobility of mercury in the environment.

Mercury and other toxic metals discharged from waste water treatment plants are associated largely with the suspended solids. Decreasing the suspended solid discharge would greatly decrease the quantity of toxic metals entering the environment at sewage outfalls.

SECTION III

INTRODUCTION

GENERAL

Prior to general recognition of mercury as an environmental hazard in the United States, most of the mercury mined, refined or imported into this country was discharged into the environment. It has been estimated[1] that of the 2340 tons of mercury used in the U.S. in 1967, about 800 tons were discharged to the atmosphere and about 650 tons to the waters. On a world basis, the corresponding estimates were 8100 tons used, 2380 tons discharged to the atmosphere, and 2250 tons discharged to the waters. Action by federal and state regulatory agencies was initially directed to industries which discharged relatively concentrated waste streams, especially the chlor-alkali industry.

Mercury occurs as a minor additive in many consumer goods and as a trace element in most raw materials. Disposal of such consumer goods and utilization of raw materials must result in discharge of large, but unknown, tonnages of mercury to the environment in relatively dilute, and thus difficult to control, waste streams. Mercury in consumer goods, and mercurials from hospitals, laboratories, and some industries, are typically brought together at municipal sewage treatment plants, and the sewage outfalls act as point sources of mercury to the waters. The imprint of such a treated sewage discharge on the sediments of Santa Monica Bay has been reported.[2]

Mercury in raw materials, especially coal and Cu-Pb-Zn ores,[1] is largely discharged to the atmosphere, from where it may re-enter the terrestrial or aquatic environments by dry fallout, or may enter the global circulation system and rain out at a location far from the point of discharge. Because of the quantities of raw materials involved, the tonnage of

mercury put into circulation may be quite large. About $3x10^9$ tons of coal are burned annually; if this coal averages 1 ppm (reported values range from 0.01 to 300 ppm) the annual mercury discharge is about 3000 tons. About 10^9 tons of Cu-Pb-Zn ores are processed annually. "Reasonable" estimates of the mercury content of these ores range from 3 to 30 ppm, corresponding to a mercury discharge of 3000 to 30,000 tons per year.

This project deals primarily with these dilute discharges of mercury into the environment. Some sources, sinks, and transport mechanisms have been explored in a preliminary way. Where feasible, studies of other metals were included.

SAMPLING AND ANALYSIS

Samples analyzed were primarily soils, sediments, natural waters, and sewage from southwestern Michigan. This area has a major advantage in that the terrain is mostly sandy, and so background levels of most metals, including mercury, are quite low - well below crustal abundances. As a consequence, the imprint of dilute discharges can be discerned much more readily than is the case in other areas of the country, where background levels are higher. Standard oceanographic sampling tools were used - teflon lined Nansen bottles and a Ponar dredge or small bottom snapper for samples from the aqueous environment, and polypropylene spoons for surface soils. In the earlier studies, wet samples were freeze-dried prior to analysis, but because of some reports of mercury losses on freeze drying later samples were analyzed as received, with the moisture content determined on a separate aliquot. Organic content of the samples was determined as loss on ignition at 550° C. Samples were initially oxidized with acidic $KMnO_4$, with final oxidation in closed tubes at 80°C with added $K_2S_2O_8$. Final quantitation was by the standard flameless AA method,[3] using the Mercometer (Anti-Pollution Technology Corporation, Holland, Michigan).

Only total mercury was determined for all samples. We were unable to develop a reliable analytical capability for organomercurials. The use of a mass spectrograph as a specific GC detector was explored, but we were unable to get adequate sensitivity for the mercury levels in our samples.

Other metals were determined by AA (Perkin-Elmer 303), using flame atomization and in a few cases, non-flame (electro-thermal) atomization.

SECTION IV

SOIL PATTERNS RESULTING FROM ATMOSPHERIC DISCHARGE

METAL DISTRIBUTIONS IN URBAN SOILS

Surface soil samples should present an integrated record of metal fallout, both dry and by precipitation. For fallout after transport over short distances, the observed distribution of metals in surface soils should be related to land use patterns. To explore this hypothesis, a study of metal distribution in soils in and around Grand Rapids, Michigan was conducted. The study area was a 19x13 mile rectangle, from which 2-inch cores of surface soils were taken on a 1 mile grid. The study area was divided into Residential, Agricultural, Industrial, and Airport zones, and the concentrations of mercury and ten other metals were correlated with land use. This study has been published[4] and some of the results are presented here in Table 1.

Most of the trace metals are enriched in the industrial zone, compared to the residential and agricultural zones. In particular, the old industrial zone, which lies along the Grand River and is contiguous with older, high density residental, is significantly enriched in the toxic metals Ag, Cd, Cr, Cu, Pb, and Zn. Mercury, and to a lesser extent cadmium, show considerably more diffuse distributions than the other metals; this may be due to a higher volatility of these elements, or to a multiplicity of sources.

Because of the proximity of many potential discharge sources, it was not usually possible to isolate point sources. In a few special cases, described below, some point sources were identified

MERCURY ENRICHMENT AROUND AN ASPHALT PLANT

The initial sampling grid showed two high values for mercury, from an

Table 1. METAL CONCENTRATIONS RELATED TO LAND USE

(Concentrations in μg of metal per gram of soil)

Metal	Residential	Agricultural	Industrial	Airport	Mean of 3 highest values
Ag	0.13	0.19	0.37	0.29	2.0
Ca	2300	1400	3200	4100	9300
Cd	0.41	0.57	0.66	0.77	2.9
Co	2.3	2.7	2.8	7.9	11
Cr	3.2	4.6	8.5	17.6	90
Cu	8.0	8.8	16.3	10.4	114
Fe	2200	2600	3100	6200	7400
Hg	0.10	0.11	0.14	0.33	1.2
Ni	5.4	5.6	8.3	12.3	38
Pb	17.9	15.4	47.7	17.9	320
Zn	21.1	22.1	56.6	36.6	280

area which is primarily undeveloped woodlands. Investigation revealed the existence of an asphalt plant in this area, and additional soil samples were taken in a grid around the plant. These soils showed a well developed fallout pattern around the plant, the shape of which correlated well with the wind rose for this area.

MERCURY ENRICHMENT AROUND AIRPORTS

The Grand Rapids Airport is relatively new, and surrounded by undeveloped and agricultural land. The soil study indicated that all metals (except lead) were concentrated around the airport, as shown in Table 1. Analysis of a sample of commercial jet fuel, JP4, did not yield detectable amounts of any of the metals found enriched, so the enrichment must be due to some other process, possibly engine corrosion. Another large, isolated airport was studied, Kincheloe Air Force Base in the eastern upper peninsula of Michigan. Although access to the base itself was restricted, samples were obtained from along minor roads bordering the base to the south, east, and west. These samples were analyzed for Hg, Cr, and Pb. South of the base, the mean values were 0.13 ppm Hg, 9.5 ppm Cr, 4.7 ppm Pb. East, the means were 0.07, 5.8, and 23.2, respectively. North, the means were 0.02, 5.0, and 20.4. The winds are predominently from the northwest. That the enrichment to the south is not due to varying vehicular traffic on the road is indicated by the values for Pb; the roadside highest in Hg and Cr is lowest in Pb, indicating low vehicle traffic on this road. We have been unable to discover the exact source of mercury enrichment, but the phenomenon appears to be real.

METAL ENRICHMENT AROUND A POWER PLANT

The Consumers Power Company plant at Port Sheldon, Michigan is in an isolated location, and acts as a point source for discharge of metals to the atmosphere. The surrounding sandy terrain has very low background levels of most metals. The distribution of mercury and nine other metals in the surface soils (upper 2 cm) surrounding this plant has been deter-

mined. The sample grid included 45 sites, with 2 samples taken from each site, from well drained, established wooded areas. The observed distribution patterns correlated well with the four-year wind rose from a nearby location. The correlation indicates that the most frequent wind component (13-18 mph) transports the metals about 6 miles. The wind rose also suggests that about 1/5 of the fallout from this plant enters Lake Michigan directly. Observed soil enrichments in Fe, Ti, Zn, Co, Cr, Cu, Ni, Cd, and Ag correlate with the quantities which are predicted assuming that the precipitator works with 90% efficiency, and that the coal burned has a composition identical to the world mean.[5] The total discharge of these metals over the 10-year life of the plant is observed to be as follows: Fe, 25,000 tons; Ti, 680 tons; Zn, 160 tons; Co, 42 tons; Cr, 35 tons; Cu, 32 tons; Ni, 30 tons; Cd, 18 tons; Ag, 0.5 tons. Assuming a mean mercury content of the coals of 0.3 ppm, about 4 tons should be found in the soils; however, only 0.05 tons were found. This indicates that mercury discharged from the burning of coal deposits by a mechanism other than short-term dry fallout, and so presumably enters a different atmospheric circulation pattern and is dispersed over a much wider area. This study has been published.[6]

SECTION V

MERCURY DISTRIBUTION IN SEDIMENTS

FLAMBEAU FLOWAGE, WISCONSIN

The Flambeau Flowage, Iron County, Wisconsin, is a region in which the fish contain elevated levels of mercury, despite the lack of any obvious source of mercury to the waters. Such situations are commonly attributed to deposition of atmospheric mercury. The mercury content of 24 fish taken from these waters ranged from 0.14 to 0.60 ppm, with a mean of 0.38.[7] A published analysis[8] of water and sediment from a single location in the lake indicated that the water was below 0.5 ppb and the sediment below 50 ppb. The lake was created by damming the Flambeau River, to flood a large area of acid soils covered primarily with tamarack vegetation. There is an outcrop of bedrock in the lake, which showed a small vein of sulfide mineralization.

For the present study 24 sediment samples were collected and analyzed for organic content, Hg, Cu, Pb, and Zn. Hg concentrations were indeed not large, averaging 0.11 ppm; the only sample containing more than 0.3 ppm was collected near the outcrop and was 0.65 ppm in Hg. The averages for the other elements were 21 ppm Cu, 11 ppm Pb, and 42 ppm Zn. If the mercury in the sediments is of natural geological origin, then the mercury levels should correlate with the levels of Cu, Pb, or Zn, with which mercury tends to be associated in ore bodies. Least squares plots of ppm Hg against ppm of the other metals all showed positive slopes, but the correlation coefficients for the plots were very poor - Hg vs Cu, 0.16; Hg vs Pb, 0.41; Hg vs Zn, 0.44. These rotten statistics, which are no doubt due to the lack of precision of our analyses at this stage in the process, as well as to the low frequency of reasonably high mercury levels (only 5 of the 24 samples had more than 0.15 ppm), prevent reaching any statistically valid conclusions concerning the geologic

orgin of the mercury.

The best correlation was found between the mercury content and the organic content, for which the least squares equation is

$$\text{ppm Hg} = 3.3\text{x}10^{-2} \text{ (\% organic)} + 0.02$$

with a correlation coefficient of 0.86. Organic content ranged from less than 1% up to 65%.

Because of the possibility that potential geochemical correlations had been distorted by mobilization of mercury from the sediments, a set of 18 surface soil samples was analyzed for the same elements. Mean concentrations were 0.03 ppm Hg, 13 ppm Cu, 5 ppm Pb, and 24 ppm Zn. Correlation coefficients between Hg and the other elements were also quite poor - Hg vs Cu, 0.28; Hg vs Pb, 0.33; Hg vs Zn, 0.34.

Thus the conclusion which is statistically most valid, but which has a considerable probability of being incorrect, is that most of the mercury in the sediments was present in the original vegetation now decomposed. We did not take alkalinity and pH measurements, but Konrad[8] states that overlying waters of low alkalinity and low pH, such as he found in this region, stimulate the accumulation of mercury by fish.

VARIOUS MICHIGAN LAKES

A total of 138 sediment samples were collected from 32 water bodies in the upper peninsula and the western lower peninsula of Michigan, as a reconnaissance effort to find any lake with high mercury in the sediments, or preferably to find a pair of lakes in similar geological environments, which might have different mercury loadings in the sediments. Klein Lake, located at Kincheloe AFB, showed the highest sediment mercury content, 0.05 ppm. Only 7 of the other 135 sediment samples had mercury levels greater than 0.02 ppm. It is nice to seek pollution and find none.

LAKE MACATAWA AND ADJACENT LAKE MICHIGAN

A study of the distribution of mercury and some other metals in the sediments of Lake Macatawa and adjacent Lake Michigan was carried out. A total of 112 surface sediment samples was collected, 84 from Lake Macatawa and 28 from Lake Michigan. Lake Macatawa is divided into two basins connected by a narrow neck. Industrial wastes and the municipal sewage effluent are discharged into the eastern basin, and nearly all of the natural drainage basin also drains into the east end of the lake. About 2/3 of the drainage basin drains into Lake Macatawa via the Black River, and about 1/4 via Pine Creek, with other sources much smaller. Natural sedimentation patterns are disturbed by annual dredging of a 20 ft. deep shipping channel, which runs the length of the lake. In addition a large carp population tends to stir up at least the fine fraction of the sediments. The western basin of Macatawa exchanges water fairly freely with Lake Michigan, especially during storms.

Lake Macatawa may be divided into seven areas: two sources - Black River and Pine Creek, the eastern and western basins, the outlet to Lake Michigan, the marinas, and the shipping channel. The results of sediment analyses for each of these areas are shown in Table 2. In nearly every case, metal concentrations are proportional to the organic content. One exception is mercury, which has its maximum value in the Pine Creek area. This creek drains only residential and agricultural (largely blueberry farms) areas, and the mercury in these sediments may result from agricultural mercurials. Nickel is high in the Black River sediments, possibly a result of metal industries upstream. The lead content of the sediments shows the strong influence of commercial and pleasure boating, being highest in the marina sediments, next highest in the shipping channel, and next highest in the west basin which is much more used for pleasure boats than is the east basin.

The 28 Lake Michigan samples were collected on a 1 mile grid, 5 miles N-S

Table 2. LAKE MACATAWA SEDIMENT ANALYSIS

(metal concentrations in µg per gram of dry sediment)

Region	% Organic	Hg	Cr	Cu	Zn	Ni	Co	Ti	Pb
Outlet to Lake Michigan	0.33	0.006	2	2	8	2	0.5	12	1
Pine Creek	1.5	0.016	7	5	33	4	1	11	3
Black River	2.7	0.005	15	15	61	21	2	21	3
West Basin	3.8	0.006	40	40	65	10	4	25	14
Marinas	8.4	0.008	60	43	130	21	6	75	28
East Basin	10.1	0.009	81	68	180	52	11	75	12
Shipping Channel	11.7	0.014	85	68	180	53	12	82	21

by 6 miles E-W. The center of the grid is a line due west of the Holland channel. Results of these analyses, given in Table 3, support the metal-organic relationship observed in Lake Macatawa.

The metal-organic relationships observed here are clearly only first-order effects which may be perturbed by such factors as nearness to sources, longshore sediment drift, deep holes which may act as sediment traps, etc. Further, since the observed percent organic correlates with the % silt and % clay, one of the latter two may be in fact the key independent variable.

Table 3. LAKE MICHIGAN SEDIMENT ANALYSES

(metal concentrations in µg per gram of dry sediment)

miles offshore	depth (meters)	Sand (%)	Silt (%)	Organic (%)	Hg	Cr	Cu	Zn	Ni	Co	Ti
1.2	16	96.5	.3	.30	0.09	3	1	9	2	2	8
2.2	19	96.6	2.0	.33	0.10	5	3	12	4	3	2
3.3	23	85.5	12.9	.76	0.11	8	3	24	4	4	20
4.3	38	70.2	25.3	1.49	0.11	16	9	81	8	4	40
5.2	40	17.9	69.0	2.92	0.16	34	17	100	8	7	50
6.3	52	6.7	91.6	4.75	0.24	62	38	160	17	9	70

SECTION VI

MERCURY IN WATERS AND SUSPENDED SOLIDS

The sediment studies suggested that mercury and other metals tend to be associated with the fine-sized fraction of the sediments, and thus that mercury is likely to be transported largely in the suspended load of rivers. Water samples were collected from the Grand River, which acts as a receiving body for large quantities of municipal and industrial wastes, and from Lake Michigan near the mouth of the Grand River. In one sampling run, 77 samples were collected from various depths at 14 stations in the Grand River and Lake Michigan. The suspended particulates were separated from the water by high-speed centrifugation, and mercury in the dissolved and suspended phases was determined. For seven of the stations the mass of suspended solid was also determined, to permit calculation of the partition factor (Hg in solid/Hg in solution). A portion of the data is presented in Table 4. Exact locations are omitted since the discharge of the river may be directed variously, depending on the Lake Michigan currents at the time of sampling. An additional set of samples, data in Table 5, was collected from the Grand River upstream from the city of Grand Rapids, and on downstream to the center of the city.

It is evident that most of the mercury in these waters is associated with the particulate matter. The suspended solids average about 10 ppm in mercury, in reasonable agreement with the values found by Cranston and Buckley[9] in Nova Scotia. This value is considerably higher than any we have found in bulk sediments of this region, demonstrating the high adsorption efficiency of the small suspended particles. The partition ratio tend to be somewhat smaller in the urban river samples; this probably is due to a difference in the particle size of the suspended solids resulting from the very high flow of the river at the time of the sampling.

Table 4. MERCURY IN SOLUTION AND SUSPENDED SOLIDS IN THE GRAND RIVER AND LAKE MICHIGAN

Location	Sample depth (ft)	Hg solution (ppb)	Hg total (ppb)	Susp. solids, (mg/ℓ)	Hg in dry solids (ppm)	$\frac{Hg(solid)}{Hg(solids)}$ ($X10^{-5}$)
1 mi. upstream	0	.045	.170	13.4	9	2
from mouth	10	.030	.170	14.4	10	3
	15	.025	.170	11.6	12	5
Mouth	0	.035	.195	12.4	13	4
	15	.035	.315	6.0	47	13
	25	.035	.095	5.6	10	3
Lake, 1 mi. W.	0	.030	.095	3.6	18	6
of mouth	30	.065	.120	2.2	25	4
	60	.030	.060	3.2	9	3
Lake, 2 mi. W.	0	.040	.100			
of mouth	40	.065	.120			
	80	.030	.060			
Lake, 7 mi. W.	0	.030	.090	2.6	23	8
of mouth	60	.025	.065	2.0	20	8
	120	.025	.080	0.8	70	28
	195	.015	.045	1.6	19	13
	240	.040	.120	1.8	44	11
Lake, 5 mi. N.W.	0	.045	.100			
of mouth	45	.030	.070			
	90	.025	.045			
	155	.025	.055			

Table 5. MERCURY IN THE GRAND RIVER IN AND UPSTREAM FROM GRAND RAPIDS

(all samples at 1 meter depth)

Location (miles upstream from city center)	Hg solution (ppb)	Hg total (ppb)	Susp. solids (mg/ℓ)	Hg in dry solids (ppm)	$\frac{\text{Hg(solid)}}{\text{Hg(sol'n)}}$ ($X10^{-5}$)
0	.080	.395	21.0	15	2
0.4	.065	.335	13.5	20	3
1.0	.040	.105	17.5	4	1
2.0	.030	.110	21.0	4	1
2.7	.045	.230	23.5	8	2
5.0	.025	.105	19.5	4	2
6.0	.040	.150	21.5	5	1
7.0	.040	.090	5.0	10	2

SECTION VII

FATE OF MERCURY IN SEWAGE TREATMENT PLANTS

Traces of mercury in consumer goods, together with mercury from hospitals and laboratories are commonly brought together in treatment plants and discharged through the outfall. Konrad[8] has reported concentrations from 0.6 to 27 ppb in influents, and from 0.5 to 3 ppb in effluents from 13 plants in Wisconsin. On occasion, the effluent concentration exceeds the influent concentration. Since an effluent concentration of 1 ppb corresponds to an annual discharge of about 1000 lb of mercury per million people served by the plant, it appears that the discharge of treated sewage adds a considerable mercury loading to the environment at the point of discharge. Elevated mercury levels in sediments around an ocean outfall have been reported[2].

The flow of mercury through the secondary treatment plants at Holland and Grand Rapids, Michigan has been studied. Samples were taken from various points in the plant, with the times adjusted so as to follow a single water mass. The analytical results and plant operating data are presented in Table 6.

In the Holland plant, each step in the treatment increased the mercury content of the effluent. This is a consequence of rather low influent mercury levels, and high sludge mercury levels. The mercury redistributes from the sludges to the stream. In the Grand Rapids plant some mercury was removed during secondary treatment.

Some studies of the partitioning of mercury and other elements between the suspended solids and solution were conducted, similar to those described in the Grand River. Partitioning ratios, expressed as (M solid/M solution) were as follows: Na-100, Mg-400, Cr-2000, Ni-4000,

Table 6. MERCURY FLOWS IN SEWAGE TREATMENT PLANTS

Sample	Holland			Grand Rapids		
	Flow (ℓ/hr)X10^{-6}	Hg ppb	Hg flow (gm/hr)	Flow (ℓ/hr)X10^{-6}	Hg ppb	Hg flow (gm/hr)
Raw	0.704	0.94	0.66	9.80	0.93	9.11
Additions to Raw	0.064(a)	34.6	2.22	0.02(b)	2.88	0.05
To Primary	0.768	4.02	3.08	9.82	0.93	9.21
Primary Sludge	0.055	42.7	2.33	0.016	17.2	0.28
Prim. Eff. to Secondary	0.714	1.09	0.78	6.16	0.91	5.61
Recycled Activated Sludge	0.965	7.37	7.11	3.16	10.08	31.85
To Secondary	1.679	4.70	7.89	9.32	3.92	36.53
Waste Act. Sludge	0.015	7.37	0.11	-0-		
Secondary Effluent	0.699	1.16	0.81	6.16	0.74	4.56
Prim. Eff. Discharged	-0-			3.63	0.91	3.30
Final Eff. Chlorinated	0.699	1.39	0.97	9.79	0.80	7.86

(a) This is a mixed heavy waste from various sludge thickening processes

(b) Filtrate from sludge thickening

Hg-7000, Al-16,000, Cu-17,000, Zn-26,000. The magnitude of these partitioning factors indicates that improved removal of these metals from waste water depends strongly on decreasing the concentration of suspended solids in the effluent.

SECTION VIII

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